

Towards Modeling Elastic Properties of Estane®

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We have, as reported earlier, developed a methodology based on self-consistent field theory by which we can determine the morphological properties of multiblock copolymer self-assemblies. We have done this in an effort to achieve modeling capabilities of morphological and elastic properties of the commercially available segmented polymer Estane®. Our interest in Estane® arises from the fact that this material acts as the polymeric binder component in the high explosive PBX 9501 and it has been strongly suggested that aging effects in this composite arise from chemical and physical changes in the binder. Estane® 5703 is a segmented block copolymer whose strength arises from the self-organization it undergoes. The chemical structure is shown in Fig. 1. The “hard” segments are comprised of methylenediphenylisocyanate (MDI) units (with 1–3 repeats), and the “soft” segments are butyleneadipate (BA) oligomers of 5 segments. These components are repeated about 28 times as shown in Fig. 1. Our self-consistent field models can effectively and precisely determine the morphological structures emerging from the self-assembly of these segmented chains.

In order to further enable this methodology to also characterize the elastic properties resulting from these self-assemblies we have constructed a self-consistent field theory that in addition to the polymer density also incorporates the local stress and strain fields in the material. The development is based on previous work by Fredrickson [1] who recently developed a self-consistent field theory for homopolymer melts which incorporates polymer density, stress, and strain fields simultaneously. The strain is included at the microscopic level via a single chain expression due to Doi and Edwards [2]. The stress field arises as the conjugate to the strain field. We have extended this theory to self-assembling polymer systems, and we have initially computer-implemented this theory for the simplest such system, namely diblock copolymers.

Within this theory we cannot only determine the morphological properties of the polymer density but also the corresponding internal stress distributions in the self-assembled structures. An example of this is shown in Fig. 2 for the hexagonal phase of a diblock copolymer. In this particular case a block copolymer composed of just two chemical species (say A and B) in the volume ratio 30:70 self-assembles into cylinders rich in the minority species (A), which are packed hexagonally in a matrix formed by the majority species (B). Together with a 2D projection of polymer density we show a similar projection of the σ_{xx} component of the stress tensor. Similarly, we can determine the remaining components of the stress tensor. Further, this model allows us to follow the local stress development in all components of the stress tensor as the self-assembly is strain

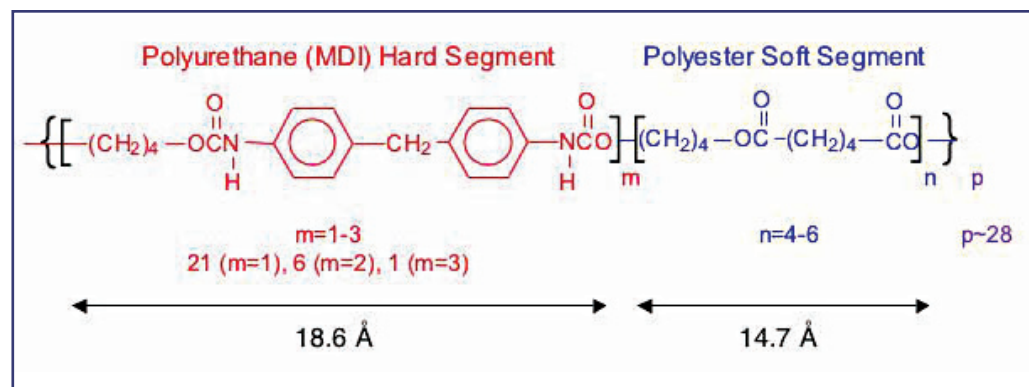
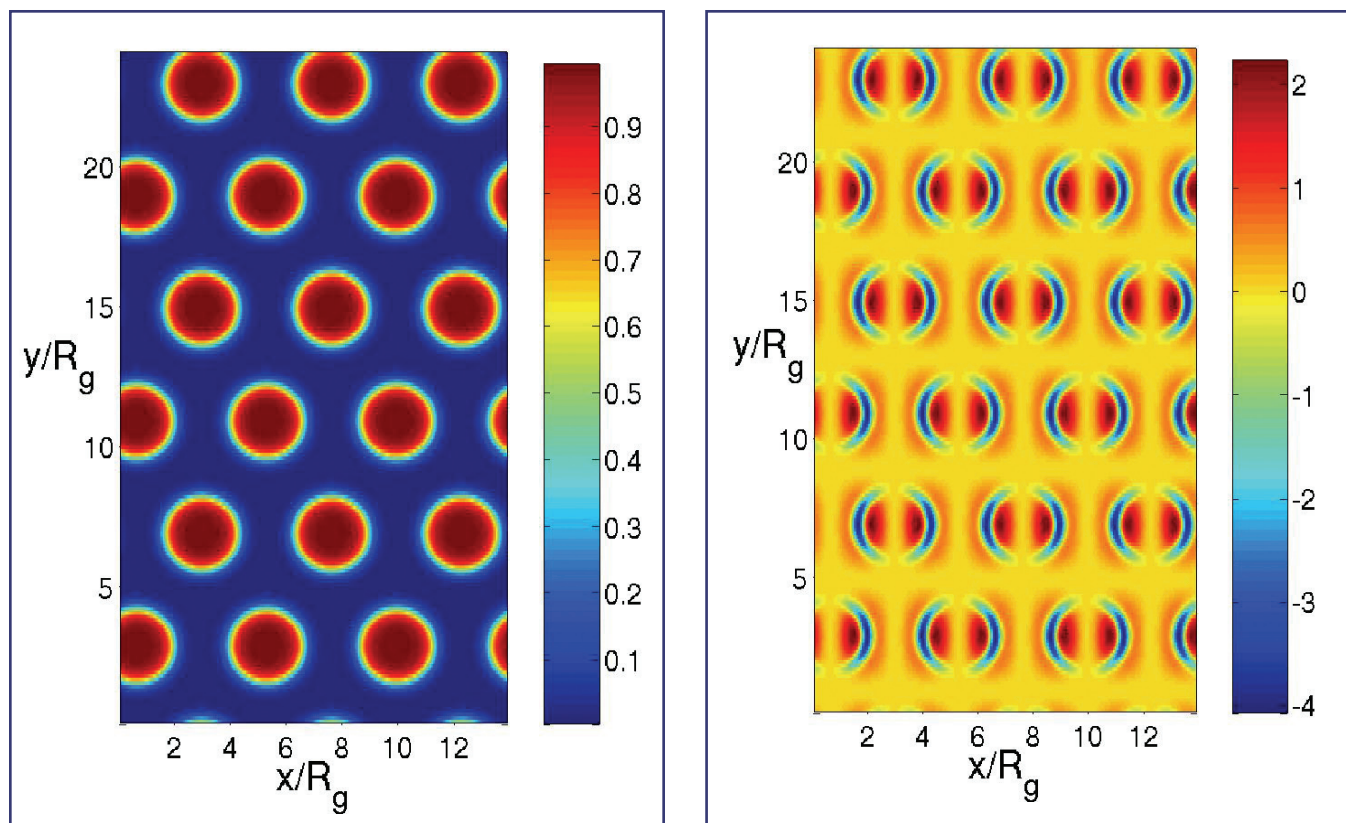


Figure 1—
The chemical structure
of Estane®.



loaded, and consequently the complete set of elastic moduli can be calculated.

We plan to apply this new technique to the more involved segmented polymer model that applies directly to Estane® and thereby help formulate a response model for this material.

[1] G.H. Fredrickson, *J. Chem. Phys.* **117**, 6810 (2002).

[2] M. Doi and S.F. Edwards, *The Theory of Polymer Dynamics* (Clarendon Press, Oxford, 1986).

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Figure 2—

Left: Two-dimensional projection of hexagonal cylindrical phase in 30:70 diblock copolymer self-assembly. Density of minority component is shown such that red indicates high density and blue low density. **Right:** The σ_{xx} component of the stress tensor corresponding to the density.